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Published as: Brennan, R.B., Healy, M. G., Grant, J., Ibrahim, T. G., Fenton, O. 2012.

Incidental phosphorus and nitrogen loss from grassland plots receiving chemically amended dairy cattle slurry. *Science of the Total Environment* 441: 132 – 140.

<http://dx.doi.org/10.1016/j.scitotenv.2012.09.078>

Incidental phosphorus and nitrogen loss from grassland plots receiving chemically amended dairy cattle slurry

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Abstract

Chemical amendment of dairy cattle slurry has been shown to effectively reduce incidental phosphorus (P) losses in runoff; however, the effects of amendments on incidental nitrogen (N) losses are not as well documented. This study examined P and N losses in runoff during three simulated rainfall events 2, 10 and 28 days after a single application of unamended/chemically

23 amended dairy cattle slurry. Twenty-five hydraulically isolated plots, each measuring 0.9 m by
24 0.4 m and instrumented with runoff collection channels, were randomly assigned the following
25 treatments: (i) grass-only, (ii) slurry-only (the study-control), (iii) slurry amended with industrial
26 grade liquid alum comprising 8% Al_2O_3 , (iv) slurry amended with industrial grade liquid poly-
27 aluminum chloride (PAC) comprising 10% Al_2O_3 , and (v) slurry amended with lime. During the
28 first rainfall event, lime was ineffective but alum and PAC effectively reduced dissolved reactive
29 P (DRP) (by 95 and 98%, respectively) and total P (TP) flow-weighted-mean-concentrations (by
30 82 and 93%, respectively) in runoff compared to the study-control. However, flow-weighted-
31 mean-concentrations of ammonium-N ($\text{NH}_4\text{-N}$) in runoff were increased with alum- (81%) and
32 lime-treated (11%) slurry compared to the study-control whereas PAC reduced the $\text{NH}_4\text{-N}$ by
33 82%. Amendments were not observed to have a significant effect on $\text{NO}_3\text{-N}$ losses during this
34 study. Slurry amendments reduced P losses for the duration of the study, whereas the effect of
35 amendments on N losses was not significant following the first event. Antecedent volumetric
36 water content of the soil or slope of the plots did not appear to affect runoff volume. However,
37 runoff volumes (and consequently loads of P and N) were observed to increase for the
38 chemically amended plots compared to the control and soil-only plots. This work highlights the
39 importance of considering both P and N losses when implementing a specific nutrient mitigation
40 measure.

41

42 **Keywords:** alum; poly-aluminum chloride; lime; runoff; amendments; management

43

44 1. Introduction

45 Incidental losses of phosphorus (P) and nitrogen (N) occur when rainfall interacts directly with
46 inorganic and organic fertilizers spread on the land surface (Preedy et al., 2001; Smith et al.,
47 2001a; Withers et al., 2003; Buda et al., 2009). Incidental P and N losses are dependent on
48 factors such as: the amount and type of fertilizer or manure applied (Kleinman and Sharpley,
49 2003), timing of the rainfall event after application of fertilizer or manure (Pote et al., 2001;
50 Smith et al., 2007; Allen and Mallarino, 2008; Hanrahan et al., 2009), the volume of runoff
51 generated, antecedent hydrologic conditions and field position, flow path length (McDowell and
52 Sharpley, 2002), vegetative cover (Zhang et al., 2003) and surface slope (Alaoui et al., 2011).
53 Incidental P losses in runoff following land application of dairy cattle slurry are dominated by
54 particulate P (PP) (Withers and Bailey, 2003) and N losses by ammonium-N ($\text{NH}_4\text{-N}$) (Smith et
55 al., 2001a). While P is generally considered the limiting nutrient in freshwater systems (Correll,
56 1998; Hudnell, 2010; Paerl, 2008; Shindler et al., 2008), N losses also pose a significant risk to
57 water quality (Johnes et al., 2007; Vitousek et al., 2009).

58

59 Chemical amendment of dairy cattle slurry (Elliot et al., 2005; Torbert et al., 2005; Brennan et
60 al., 2011a, b) and poultry litter (Moore and Edwards, 2007) has been effective at reducing P
61 losses in surface runoff following land application. As a result, manure amendment is a
62 recommended best management practice (BMP) in the USA, and federal support is available to
63 aid its implementation (Sharpley et al., 2006; SERA-17, 2012; USDA-NRCS, 2012). There have
64 been a large number of laboratory-scale studies that have examined the effect of amendments on
65 P solubility in dairy and swine slurry (Dao, 1999; Dao and Daniel, 2002; Dou and Cavigelli,
66 2003; Torbert et al., 2005). Torbert et al. (2005) amended composted dairy manure with ferrous
67 sulphate, gypsum and lime (each at 3:1 metal-to-total phosphorus (TP) ratio) before surface

68 application and immediately prior to a 40-min overland event equivalent to a rainfall intensity of
69 12.4 cm h⁻¹. Ferrous sulphate reduced dissolved reactive phosphorus (DRP) loss by 66.3%, while
70 gypsum and lime amendments increased DRP loss. In a plot study, Smith et al. (2001b) amended
71 swine manure with alum and aluminum chloride (AlCl₃) at two stoichiometric ratios (0.5:1 and
72 1:1 Al: TP). Dissolved reactive phosphorus reductions for alum and AlCl₃ at the lower ratio were
73 33% and 45%, respectively, and 84% for both amendments at the higher ratio.

74

75 While the effectiveness of amendments is well established, there is less information on the effect
76 of amendments on N loss to runoff and runoff properties. It is known that land application of
77 dairy cattle slurry on grassland (Nunez et al., 2001) and arable land increases runoff volumes
78 which affects N and P losses (Smith et al., 2007). In addition chemical amendment of dairy cattle
79 slurry affects the texture and rate of drying of slurry following land application (Brennan et al.,
80 unpublished data) which may impact runoff volumes. Approximately 50% of the N in dairy
81 cattle slurry is in an inorganic form (NH₄-N from urea in the urine component of slurry) and
82 although this is plant available, as much as 80% of it is lost through volatilization in a short time
83 period after slurry application. Chemical amendments have been shown to significantly reduce
84 ammonia (NH₃) volatilization following land spreading of dairy cattle slurry (Lefcourt and
85 Meisinger, 2001). This is likely to increase the NH₄-N available for uptake by plants and
86 potentially runoff.

87

88 Chemical amendments reduce P solubility in poultry, swine and dairy cattle manure. However,
89 slurry N is much more mobile than P and its loss pathways are more complex. Therefore,
90 amendments which change the properties of slurry may influence N transformations following

91 land application, and may result in increased N losses to the atmosphere or in surface runoff.
92 This is sometimes referred to as ‘polluting swapping’ (Stevens and Quinton, 2009). Therefore,
93 any study investigating the efficacy of any potential P mitigation measure, such as those
94 described above, must also consider the ‘pollution swapping’ that may arise from their use. To
95 the authors’ knowledge, this is the first study to examine the impact of chemical amendment of
96 dairy cattle slurry on incidental losses of both N and P in runoff.

97

98 The specific objectives of this study were to investigate (i) incidental N and P losses from soil-
99 only, slurry-only and amended slurry treatments (ii) the effect of chemical amendment of dairy
100 cattle slurry on runoff volume, volumetric water content, and time to runoff, and (iii) the short-
101 term effect of land application of chemically amended dairy cattle slurry on soil chemical
102 properties.

103

104 **2. Materials and Methods**

105

106 **2.1. Study site characterization**

107

108 The site work was carried out between 11th September 2010 and 18th October 2010, on a 0.6-ha
109 isolated plot on a beef farm located at Teagasc, Johnstown Castle, Environmental Research
110 Centre (latitude 52° 17’N, longitude 6° 29’W), in the southeast of Ireland. This area has a cool
111 maritime climate, a mean annual precipitation of 1002 mm (effective rainfall (rainfall -
112 evapotranspiration) from between 400 to 500 mm), and a mean annual temperature of 10°C
113 (Ryan and Fanning, 1996).

114

115 The location of 25 isolated plots within the 0.6 ha site was determined by: topography/slope, soil
116 texture/drainage assessment, depth to watertable, and soil nutrient analysis. Within the 25 plots
117 (0.9 m by 0.4 m), treatments were randomly assigned in five blocks (Fig 1). The site had
118 undulating topography with a 6.7% slope along the length of the site and an average slope of
119 3.6% across the site. For textural analysis (pipette method, B.S.1377-2:1990 (BSI, 1990)), 10
120 cm-deep soil samples ($n=3$) were taken from a 1-m² area at the top, middle and bottom of the 0.6
121 ha plot (Fig 1). Electromagnetic conductivity (characterization to 4 m below ground level (bgl))
122 and resistivity of the 0.6-ha site were used to infer overall textural and drainage characteristics.
123 The top of the plot comprised gravelly clay with pockets of silty/clayey gravel underlain by
124 silt/gravel (20 to 26 mS m⁻¹), and was relatively well-drained compared to the lower part of the
125 site, which comprised silt/clay and was poorly drained (>26 mS m⁻¹). The median perched
126 watertable depth in three piezometers (top, middle and bottom of slope) was 0.6 m bgl on site.
127 The nutrient status of the soil at these locations (P, potassium (K), and magnesium (Mg)),
128 determined using Morgan's extractant (Morgan, 1941), are presented in Table 1. Soil pH ($n=3$)
129 was determined using a pH probe and a 2:1 ratio of deionised water to soil (Table 1). Each plot
130 was installed, isolated and instrumented with a runoff collection channel (Fig 1). A composite
131 soil sample (100 mm) was taken from each plot (before (t_0) and after the experiment (t_{30})) and
132 soil pH, Morgan's P, K, Mg and lime requirement (LR) were determined. In addition, composite
133 soil samples (25 mm) were taken from each plot at t_0 and t_{30} for water extractable P (WEP)
134 determination.

135

136 2.2. Slurry analysis

137

138 Dairy cattle slurry was collected from the dairy farm at the Teagasc, Environmental Research
139 Centre, Johnstown Castle, in September of 2010. The storage tanks were agitated and slurry
140 samples were transported to the laboratory in 25-L drums. Slurry samples were stored at 4°C
141 prior to land application. Slurry pH was determined using a pH probe (WTW, Germany). The TP
142 of the dairy cattle slurry was determined after Byrne (1979). Total potassium (TK), total nitrogen
143 (TN) and TP were carried out colorimetrically using an automatic flow-through unit (Varian
144 Spectra 400 Atomic Absorption instrument). The WEP of slurry and amended slurry was
145 measured at the time of land application (1:100 dry matter slurry: deionised H₂O) after Kleinman
146 et al. (2007), and NH₄-N of slurry and amended slurry was extracted by shaking 50 g of slurry in
147 1 L of 0.1 M hydrochloric acid (HCl) on a peripheral shaker for 1 hr and filtering through No. 2
148 Whatman filter paper at the time of application. The results of the slurry analysis are shown in
149 Table 2. The slurry used in this study was typical of slurry found on farms in Ireland (Fenton et
150 al., 2011). The slurry TN, TP, NH₄-N and TK were constant across samples. The WEP of slurry
151 was decreased significantly by all alum and PAC amendments. Alum addition reduced the slurry
152 pH from approximately 7.1 to 6.5, while lime addition increased the slurry pH to 8.8.

153

154 **2.3. Treatments**

155

156 The five treatments examined in this study were (i) grassed soil-only (referred to as soil-only
157 hereafter) (ii) slurry applied to grassed soil (the study-control) (iii) slurry amended with
158 industrial grade liquid alum (Al₂(SO₄)₃.nH₂O), comprising 8% Al₂O₃ (iv) slurry amended with
159 industrial grade liquid PAC (Aln(OH)mCl₃n-m), comprising 10% Al₂O₃, and (v) slurry amended

160 with lime ($\text{Ca}(\text{OH})_2$). The slurry and amendments were mixed by shaking in 2-L containers for
161 30 s immediately prior to land application. In practice, it is likely that amendments would be
162 mixed with the slurry in storage tanks during slurry agitation, which normally occurs within 24 h
163 of land application. Two days before the first rainfall simulation, slurry and amended slurry were
164 applied directly to the surface of the grassed soil. Slurry application rates were equivalent to 33
165 m^3 slurry ha^{-1} (42 kg TP ha^{-1}), the rate most commonly used in Ireland (Coulter and Lalor, 2008).
166 Amendments were applied at stoichiometric ratios determined based on results of Brennan et al.
167 (2011b). Alum was applied at a rate of 1:1 (Al: TP); PAC at a rate of 0.85:1 (Al: TP); and lime at
168 a rate of 3.9:1 (Ca:TP). Land application of treatments was staggered over three days and applied
169 in blocks to allow for the first rainfall event (RS1) two days after land application of slurry.

170

171 **2.4. Rainfall event simulation and plot design**

172

173 Two identical portable multi-drop ‘Amsterdam type’ rainfall simulators, described by Bowyer-
174 Bower and Burt (1989), were used in this study. These rainfall simulators have been used on
175 similar permanent grassland sites and soil types (Kurz et al., 2006; Kramers et al., 2009;
176 O’Rourke et al., 2010). The rainfall simulators were designed to distribute rainfall over a surface
177 area of 0.5 m^2 and were calibrated to deliver rainfall at an intensity of 11 mm hr^{-1} . The rainfall
178 simulator water had average concentrations for the three rainfall simulation events of 0.05 mg
179 $\text{NH}_4\text{-N L}^{-1}$, $4.61 \text{ mg nitrate-N (NO}_3\text{-N) L}^{-1}$, $0.002 \text{ mg DRP L}^{-1}$ and $0.004 \text{ mg TP L}^{-1}$.

180

181 In order to ensure the absence of edge effects, the rainfall simulators were located directly above
182 study plots – each measuring 0.36 m^2 in area. The plots were isolated using 2.2 m-long, 100 mm-

183 deep rigid plastic sheets, which were pushed 50 mm into the soil to isolate three sides of the plot.
184 The runoff collection channel was placed at the bottom of the slope (Fig 1). Plots were orientated
185 with longest dimension in the direction of the slope (average 3.6%). The runoff collector
186 comprised a polypropylene plastic U-shaped channel piece, which was cut in half and wedged
187 against the soil at a depth of approximately 25 mm below the soil surface (Fig 1). A 400 mm-
188 wide edging tool was used to cut the soil to ensure a good seal between soil and collector. The
189 plots were left uncovered for two weeks prior to first rainfall simulation to allow natural rainfall
190 to wash away soil disturbed by inserting the isolators. Natural rainfall was excluded from the
191 plots between time of slurry application and RS1. Thereafter, plots were exposed to natural
192 rainfall. Natural rainfall, together with the average simulated rainfall applied for each of the
193 rainfall simulations, is shown in Fig 2. The grass on all plots was clipped to a height of 50 mm
194 two days prior to application of treatments to simulate the spreading of slurry following silage
195 cutting, which is common practice in Ireland. The second rainfall event (RS2) was 10 days after
196 the original application ($t = 12$ d) and the third (RS3) after 28 days ($t = 30$ d).

197

198 Soil Moisture deficit (SMD) for the entire landscape position was estimated using the grassland
199 Hybrid model of Schulte et al. (2005). For all events, rainfall simulator amounts (mm) were
200 added to actual daily rainfall data and the SMD for each subsequent day was estimated (based on
201 well, moderately and poorly drained soil). When SMD values returned to values achieved using
202 actual rainfall data, the subsequent simulated rainfall event took place. The volumetric water
203 content of soil in each plot was measured immediately prior to each rainfall simulation event
204 using time domain reflectrometry (Delta-T Devices Ltd., Cambridge, UK), which was calibrated
205 to measure resistivity in the upper 50 mm of the soil in each plot.

206

207 **2.5 Runoff sample collection and analysis**

208

209 Surface runoff was judged to occur once 50 ml of water was collected from the runoff collection
210 channel and the time from start of rainfall simulation to runoff of 50 ml being the time to runoff
211 (TR). Samples were collected every 5 min for RS1, and every 10 min for RS2 and RS3. Surface
212 runoff was collected for 30 min once runoff commenced until the rainfall simulator was switched
213 off to allow the flow-weighted mean concentration (FWMC) to be calculated (Kurz et al., 2006).
214 For the third rainfall event, water was sprayed gently on the plots using a watering can until
215 surface ponding occurred in order to complete rainfall simulations in daylight hours.

216

217 Immediately after collection, runoff water samples were filtered through 0.45 µm filter paper and
218 a subsample was analyzed colorimetrically for DRP, NO₃-N, NO₂-N and NH₄-N using a nutrient
219 analyzer (Konelab 20, Thermo Clinical Labsystems, Finland). A second filtered subsample was
220 analyzed for total dissolved phosphorus (TDP) using acid persulphate digestion. Unfiltered
221 runoff water samples were analyzed for TP with an acid persulphate digestion. Particulate
222 phosphorus was calculated by subtracting TDP from TP. The DRP was subtracted from the TDP
223 to give the dissolved un-reactive phosphorus (DUP). All samples were tested in accordance with
224 the Standard Methods (APHA, 2005).

225

226 **2.6 Data analysis**

227

228 Runoff ratio (RR) for each plot and for the duration of each simulated rainfall event was
229 calculated by dividing the amount of water generated in overland flow by the amount of rainfall
230 applied. As the plots were the same size, there was no scale effect (Wainwright and Parsons,
231 2002; Norbiato et al., 2009). Differences in RR between plots result from differences in soil
232 permeability (Norbiato et al., 2009) (runoff ratio increases with a decrease in permeability), slope
233 (Alaoui et al., 2011) (increasing slope will increase RR) and depth of unsaturated zone. A higher
234 RR results from wetter rainfall pre-events and/or rainfall event conditions.

235

236 The structure of the data set was a blocked one-way classification (treatments) with repeated
237 measures over time (rainfall events (RS1-RS3)). The analysis was conducted using Proc Mixed
238 in SAS software (SAS, 2004) with the inclusion of a covariance model to estimate the correlation
239 between rainfall events. A large number of covariates were recorded, including measurements on
240 the simulators and for each analysis; this set of covariates was screened for any effects that
241 should be included in an analysis of covariance. The interpretation was conducted as a treatment
242 by time factorial. Comparisons between means were made with compensation for multiple
243 testing effects using the Tukey adjustment to p-values. Significant interactions were interpreted
244 using simple effects before making mean comparisons. In order to ensure that variation did not
245 affect the experiment, STP was included as a variable in the statistical analysis. Slurry
246 concentration, which was of much greater significance in terms of P concentrations in runoff
247 following slurry application, was uniform within each block.

248

249 **3. Results**

250

3.1 Incidental nutrient losses over three rainfall events

The FWMC and total loads of DRP and TP for all treatments over the three rainfall simulation events are presented in Fig 3. Slurry application increased the FWMC and total loads of DRP and TP. Alum and PAC were equally effective at reducing FWMCs of DRP and TP compared to the study-control. Lime amendment resulted in increased FWMCs of DRP and TP compared to the study-control, with total loads for the lime treatment approximately 2 times greater than for slurry DRP and TP. When total loads were considered, PAC performed better than alum in reducing total loads of DRP. The effects of amendments on P loss were not significant for RS2 and RS3, which is likely a result of available P being leached from the soil.

The FWMC and total loads of $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ for all treatments are presented in Fig 4. The addition of alum resulted in an increase in the FWMC of $\text{NH}_4\text{-N}$ compared with the study-control, while both lime and PAC treatments decreased the $\text{NH}_4\text{-N}$ loss. In contrast, all amendments resulted in an increase in the FWMC of $\text{NO}_3\text{-N}$ compared with the study-control. The PAC amendment was the only amendment which decreased total loads of $\text{NH}_4\text{-N}$ to below those of the study-control. In contrast, both alum and lime amendments resulted in an increase in $\text{NH}_4\text{-N}$ loads compared with the slurry treatment. Nitrite losses were negligible and were equivalent to approximately 1.9% of NO_3 for all samples and, for this reason, were not plotted in Fig 4.

3.2 Runoff characteristics

274 The time from start of rainfall simulation event to commencement of runoff event is shown in
 275 Fig 5. Time to runoff was generally longer for RS2 and shorter for RS3 (pre-wetted plots). No
 276 clear patterns were observed between treatments and differences were not significant. Total
 277 runoff volumes for the study were similar for soil and alum treatments (3990 ml 3930 ml), lower
 278 for the slurry treatment (3670 ml) and higher for lime and PAC treatments (4780 ml and 4460
 279 ml). The differences observed between treatments were not statistically significant. There was no
 280 experimental effect on TR across all treatments when rainfall and rainfall intensity were included
 281 as covariates in the model. Both covariates showed a quadratic effect. Although there were no
 282 treatment effects observed for volumetric water content (VMC), RR and volume runoff,
 283 significant event effects were observed. Antecedent SMD conditions before all rainfall
 284 simulations for different drainage classes are presented in Fig 2. Soil moisture deficit was similar
 285 for all three rainfall events.

286

287 3.3 Soil test P, K, LR and pH

288

289 Soil test P, WEP, Mg, K, pH and LR results from analysis of plots before (t_0) and at the end of
 290 the experiment (t_{30}) are presented in Table 3. Average STP, Mg and K concentrations before the
 291 start of the experiment were similar for soil (5.5, 182 and 58 mg L⁻¹), slurry (4.5, 173 and 57 mg
 292 L⁻¹) and amended plots (from 4.3 to 5.9 mg L⁻¹, from 160 to 194 mg L⁻¹ and from 53 to 59 mg L-
 293 ¹). At the end of the experiment, STP increased by 13% in soil-only plots and by 28 to 34% in
 294 slurry, PAC and alum. Lime showed an 8.8% decrease in STP. At the end of the experiment, soil
 295 K increased for all treatments. Soil WEP decreased between t_0 and t_{30} for soil-only, alum-

296 amended and PAC plots (20, 4 and 37%) and increased for study-control and lime-amended plots
297 (42 and 64%).

298

299 **4. Discussion**

300

301 Under the European Union (EU) Water Framework Directive (WFD) (EU WFD; 2000/60/EC,
302 OJEC, 2000), the water quality of surface and ground waters should be of ‘good status’ by 2015.

303 Small amounts of P losses may contaminate large quantities of water and, therefore, incidental
304 losses are of concern, in particular, for flashy events during baseflow conditions. Chemical
305 amendment of dairy slurry has been shown to be effective in this regard. Moving from laboratory
306 to field scales allows incidental losses to be simulated using *in-situ* soil and drainage conditions.

307 The impact of slurry and amended slurry on soil pH, infiltration and runoff volumes,
308 concentrations and loads, are all important when assessing the feasibility of a particular
309 amendment.

310

311 **4.1 Incidental losses for all rainfall events**

312

313 In order to assess the adverse effects of discharge of incidental losses to a surface waterbody, it is
314 critical to examine both runoff nutrient concentrations and total loads. Statistical analysis showed
315 that differences in runoff volume between treatments were not significant. The addition of lime
316 to soil or slurry, which is applied directly to soil, can change soil hydraulic characteristics such
317 as infiltration, water retention and hydraulic conductivity, and may lead to lower (Roth and
318 Pavan, 1991) or higher (Tarchitzky et al., 1993) runoff volumes. The increase in P loss as a result

319 of lime amendment may be also due to an increase in the pH of the lime-amended slurry. Penn et
320 al. (2011) found that in order for calcium (Ca)-phosphate bonds to remain stable, the pH must
321 remain in a range of 6.5 to 7.5. In the present study, the average pH of the soil on the study site
322 was 6.0 and the pH of the lime-amended slurry was 8.8 at the time of application. Brennan et al.
323 (2011a) showed that the pH of lime-amended dairy cattle slurry increased in the first 24 hr
324 following land application. The slurry pH was too high for Ca-P bonds to be stable during RS1
325 and when the slurry and soil interacted and reached equilibrium, the soil pH was lower than the
326 optimal pH for the formation of Ca-P bonds. This may explain why reductions were not observed
327 during RS2 and RS3. In the Brennan et al. (2011b) study, lime was applied at 10:1 Ca:TP
328 compared to 3.9:1 in the present study, and this is possibly the reason for the difference in
329 performance. In addition, the soil used in the Brennan et al. (2011b) study had a pH of 7.45
330 compared to 5.94 in the present study.

331

332 The reductions achieved in this study are consistent with the findings in Brennan et al. (2011b)
333 with alum being the most effective amendment at reducing incidental PP and TP losses, while
334 PAC was most effective at reducing DRP losses. Incidental P losses accounted for the majority
335 of P losses from the study-control plots, with approximately 75% of DRP, 72% of DUP, 94% of
336 PP and 83% of TP losses, measured over the three rainfall events, occurring during RS1. While
337 incidental losses were significantly reduced in the alum and PAC-amended plots, the effect of
338 amendments on chronic loss of P from the plots was not clear, as differences in runoff
339 concentrations during RS2 and RS3 were not statistically different to the study-control. Studies
340 have shown that chemical amendments can reduce incidental and chronic P losses (long-term P
341 losses to runoff arising from elevated STP (Buda et al., 2009)) from soils receiving amended

342 poultry litter (Moore and Edwards, 2005 and 2007). Amendments must be an ongoing practice
343 for every manure application to effectively reduce P losses. Ultimately, P application must be
344 balanced with crop P requirements to avoid chronic P loss.

345

346 In the present study, chemical amendment of dairy cattle slurry had no significant effect on NO_3^- -
347 N concentration or load in runoff water. Alum increased the FWMC and load of $\text{NH}_4\text{-N}$
348 compared to the study-control during the first rainfall event, PAC reduced the FWMC and load
349 of $\text{NH}_4\text{-N}$ and lime had no effect on the FWMC but increased the load of $\text{NH}_4\text{-N}$ due to an
350 increase in runoff volume. Dairy cattle slurry is high in $\text{NH}_4\text{-N}$ which explains the high $\text{NH}_4\text{-N}$
351 in runoff during RS1 (Smith et al., 2007). In a gas chamber experiment, Brennan et al.
352 (unpublished data), using the same amendments as Brennan et al. (2011b), found that alum and
353 PAC reduced NH_3 emissions from land applied slurry by up to 93% while lime amendment
354 resulted in a two-fold increase in NH_3 emissions. The increase in $\text{NH}_4\text{-N}$ load observed for the
355 alum treatment during RS1 was likely caused by a decrease in NH_3 volatilization, which resulted
356 in more $\text{NH}_4\text{-N}$ remaining on the soil surface and being available for uptake by runoff. The
357 difference between alum and PAC treatments indicates that PAC maybe more effective at
358 binding $\text{NH}_4\text{-N}$ which has not been volatilized on the soil surface, thereby reducing loss to
359 runoff. The reduction in $\text{NH}_4\text{-N}$ concentrations in runoff between RS1 and RS2 across all
360 treatments, including the study-control, was likely due to nitrification occurring in the soil
361 following slurry application and interaction with the soil. Smith et al. (2007) added dairy cattle
362 slurry at a rate $75 \text{ m}^3 \text{ ha}^{-1}$ to grassed plots and reported soluble N ($\text{NH}_4\text{-N} + \text{NO}_3\text{-N}$)
363 concentrations ranging from 2 mg L^{-1} to 14 mg L^{-1} , which was comparable to the average
364 FWMC of soluble N observed in the present study (6.3 mg L^{-1}). The results of the present study

365 results suggest that PAC is the most suitable amendment, as there was no increase in N losses
366 compared to the study-control. This study did not examine the effect of amendments on N
367 leaching losses. This work highlights the need to examine the pollution swapping effects of all P
368 mitigation practices.

369

370 **4.2 Runoff characteristics**

371

372 In the current study, differences in slope of plots were not shown to be significant. All plots had
373 the same landscape position mid-way between a down-gradient river and an up-gradient
374 groundwater divide. Other studies have shown greater differences in slope at different landscape
375 positions. Kleinman et al. (2006) investigated P and N losses in runoff from 1 x 2 m plots under
376 simulated rainfall conditions during wet and dry periods in two landscape positions, foot slope
377 (6%) and mid-slope (30%). Kleinman et al. (2006) showed that antecedent soil moisture at the
378 foot-slope during the spring resulted in quicker runoff generation times and greater volumes of
379 runoff.

380

381 In a homogeneous soil, runoff ratios should increase with VWC. The fact that this relationship
382 was not always found in the current study for soil-only plots may be due to local heterogeneity.
383 After slurry application, this relationship was more evident, which infers that mixing of soil and
384 slurry leads to greater spatial homogeneity of water distribution and saturation. For amended
385 slurry, the higher variability between VWC and runoff ratio (often a variable relationship)
386 suggests that the amendments had a sealing effect. Within the timeframe of this study, it was not
387 possible to assess the long-term effect of amendments on soil physical characteristics. As time

388 from slurry application increases, soil conditions will return to a more heterogeneous state,
389 whilst amendments may delay this process.

390

391 **4.3 STP, K, LR and pH**

392

393 In the present study, observed differences in soil nutrient concentrations following chemical
394 amendment were not statistically significant. There were, however, noticeable changes in soil pH
395 for some plots. These changes identify a need to examine the effect of chemical amendments on
396 long-term P dynamics in soil following application of chemically amended dairy cattle slurry.

397 Studies to date involving chemical amendment of dairy slurry have largely focused on reducing

398 P solubility in dairy cattle slurry (Dao and Daniel, 2002; Dou et al, 2003; Brennan et al., 201 1a)

399 and mitigating incidental P losses in runoff studies (Smith et al., 2001b; Elliot et al, 2005;

400 Torbert et al., 2005; Brennan et al., 201 1b), but little attention has been given to the effect of

401 chemical amendments on short and long-term nutrient availability to plants. In the US, where

402 chemical amendment of poultry litter is a BMP, Moore and Edwards (2005) and Moore and

403 Edwards (2007) reported results from a 20-year study, which began in 1995 and examined the

404 effects of chemical amendment of poultry litter on soil productivity and water quality. They

405 found that long-term land application of alum-amended poultry litter did not acidify soil in the

406 same way as $\text{NH}_4\text{-N}$ fertilizers, long-term P losses were reduced, and Al availability was lower

407 from plots receiving alum-treated poultry manure than $\text{NH}_4\text{-N}$ fertilizer.

408

409 With the exception of Kalbasi and Karthikeyan (2004), there has been little research on the effect

410 of land spreading of chemically amended dairy cattle slurry to soil. Kalbasi and Karthikeyan

(2004) examined three silt loam soils with different STPs (12, 66 and 94 mg kg⁻¹ Bray-1 P, respectively) in an incubation experiment conducted over a 24-mo period. Kalbasi and Karthikeyan (2004) found that alum and ferric chloride had no effect on soil pH, while lime increased soil pH slightly. This was consistent with the findings of the present study. These results were also consistent with another study by Brennan et al. (unpublished data). In that study, 5 soils, including soil taken from the same study site as the present study, were amended with chemical amendments and incubated for 9 months. While chemical amendments consistently reduced WEP, the STP and soil pH were not significantly affected by application of amended slurry, with the exception of FeCl₃-amended slurry in some instances. Due to the relatively short duration of the present study, it was not possible to examine the relationship between the STP of incubated soils and the *in-situ* STP when subject to a similar treatment.

4.4 Management implications of using chemically amended dairy slurry

Ireland has committed to meeting the requirements of the WFD to achieve at least ‘good status’ of all surface and groundwater by 2015. While current practices are effective, there will be a time-lag before current changes in farming practices will result in an observable reduction in nutrient losses and a reduction in risk to water quality. The time-lag will be site-specific and while it is likely that in many areas the effects will be shown relatively quickly, there may be a need for some new P mitigation measures. Results show that chemical amendments can significantly reduce P losses and that a once-off application of any of the chemical amendments examined will not result in a significant change in soil physical and chemical properties. It is,

433 however, critical that the long-term effect of repeated applications of chemical amendments to
434 slurry on STP, soil pH, soil WEP, soil microbiology and macro-biology be examined.

435

436 **5. Conclusions**

437

438 The findings of this study validate findings at laboratory-scale, with amendment of dairy cattle
439 slurry with alum and PAC reducing DRP and TP losses (FWMC and loads) compared to the
440 study-control. Alum was the most effective amendment at reducing PP and TP losses, while PAC
441 was the most effective at reducing DRP losses. This study also showed that chemical amendment
442 of dairy cattle slurry with alum increased $\text{NH}_4\text{-N}$ loss (FWMC and loads) to runoff, while PAC
443 reduced $\text{NH}_4\text{-N}$ losses. Future work must examine the effects of chemical amendment of dairy
444 cattle slurry on the N cycle and gaseous emissions. In addition, these results indicate that
445 amendments may affect runoff volume for events occurring 48 hr after slurry application.
446 Following from this study, the next step will be to examine the targeted use of chemical
447 amendments at field and catchment-scale. In future, farm nutrient management must focus on
448 examining all farms within a catchment and identifying areas which pose the greatest risk. It is
449 possible that P mitigating methods, such as chemical amendment of dairy cattle slurry, may be
450 used strategically within a catchment to bind P in cow and pig slurries. This work highlights the
451 importance of considering both P and N losses when implementing a specific nutrient mitigation
452 measure.

453

454 **Acknowledgments**

455

456 The first author gratefully acknowledges the award of a Walsh Fellowship by Teagasc to support
457 this study. The authors are also grateful for assistance provided by Teagasc and NUI Galway
458 staff and colleagues with special mention to Peter Fahy, Theresa Cowman, Carmel O'Connor,
459 Denis Brennan, Linda Finn, Paddy Sills, Pat Donnelly, Stan Lalor, Nyncke Hoekstra and Paddy
460 Hayes. The authors would also like to thank Michael Brennan for his assistance with fieldwork.

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Table 1 Soil pH, Morgan's extractable P, K and Mg, sand silt, clay factions, textural class of soil within 0.6 ha plot.

Position mgL ⁻¹	Piezometer No. ¹	pH	Morgan's P	Pindex ²	K mgL ⁻¹	Mg mgL ⁻¹	Sand %	Silt %	Clay %	Textural Class
Lower	1	5.8	2.6	2	173	171	52	30	18	Sandy Loam
Middle	2	5.9	3.2	3	140	195	47	36	18	Sandy Silt Loam
Upper	3	6.1	3.6	3	96	151	44	36	21	Clay Loam
Average		5.9	3.1		136	172	47.7	34.0	19.0	
Stddev		0.2	0.5		38.6	22	4	3.5	1.7	

¹The location of the piezometers is illustrated in Fig 1.

²P Index 2 expects a likely response to fertilizers whereas a P index of 3 expects a tenuous or unlikely response.

704 Table 2 Slurry DM, pH, water extractable phosphorus (WEP), total nitrogen (TN), total phosphorus (TP) and total
705 potassium (TK) and average concentrations of NH₄-N (n=5).
706

Treatment	DM	pH	WEP	TN	TP	TK	NH ₄ ⁺ -N
			g kg ⁻¹	mg L ⁻¹	mg L ⁻¹	mg L ⁻¹	mg L ⁻¹
Slurry (control)	9.1 (0.54)	7.1 (0.62)	3.19 (0.37)	3960 (741)	1240 (145)	5170 (870)	1200 (260)
Alum	9.6 (0.58)	6.5 (0.44)	0.003(0.001)	4410 (590)	1260 (190)	5210 (640)	1160 (270)
PAC	9.42 (0.64)	6.9 (0.47)	0.007 (0.008)	3980 (1280)	1200 (270)	4330 (1290)	1180 (290)
Lime	9.4 (0.38)	8.8 (0.67)	2.48 (0.99)	5010 (725)	1390 (150)	5610 (840)	1210 (300)
Average	9.38	7.325	1.4	4340	1270	5080	1190
Stddev	0.2	1.01	1.7	492	82.2	538	22.2

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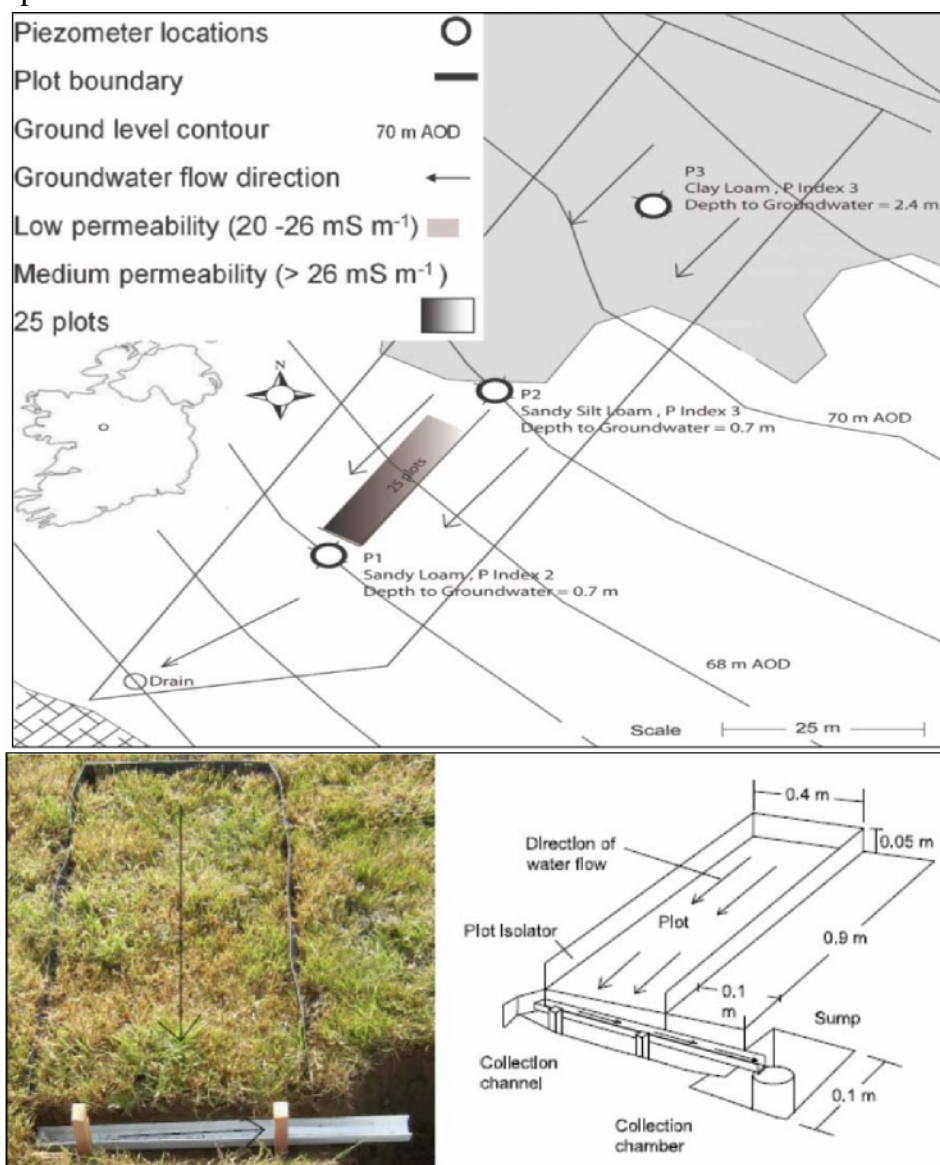
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Table 3 The average slope, soil pH, soil water extractable P (WEP), Morgan's extractable P, potassium (K), magnesium (Mg) and lime requirement (LR) on the day before the experiment (t_0) and after the experiment (t_{30}) for all of the treatments.

Treatment	Slope	pH ₀ /pH ₃₀	WEP ₀ /WEP ₃₀	P ₀ /P ₃₀	K ₀ /K ₃₀	Mg ₀ /Mg ₃₀	LR ₀ /LR ₃₀
	%		mgkg ⁻¹	mg L ⁻¹	mg L ⁻¹	mg L ⁻¹	mg L ⁻¹
Soil	3.05	5.97/5.97	4.13/6.32	5.78/3.86	57.9/93.4	182/180	4.88/4.38
Slurry (control)	2.90	5.82/5.97	8.11/4.78	6.31/8.98	56.8/91.0	173/186	6.00/4.20
Alum	4.38	5.92/5.83	6.17/6.77	5.03/5.26	52.9/66.9	194/192	5.30/5.00
Lime	3.75	6.06/6.04	8.82/9.71	6.82/11.22	59.1/80.0	188/199	4.20/3.80
PAC	3.68	5.93/6.11	6.99/5.17	6.99/5.12	58.6/93.7	160/193	5.10/3.30

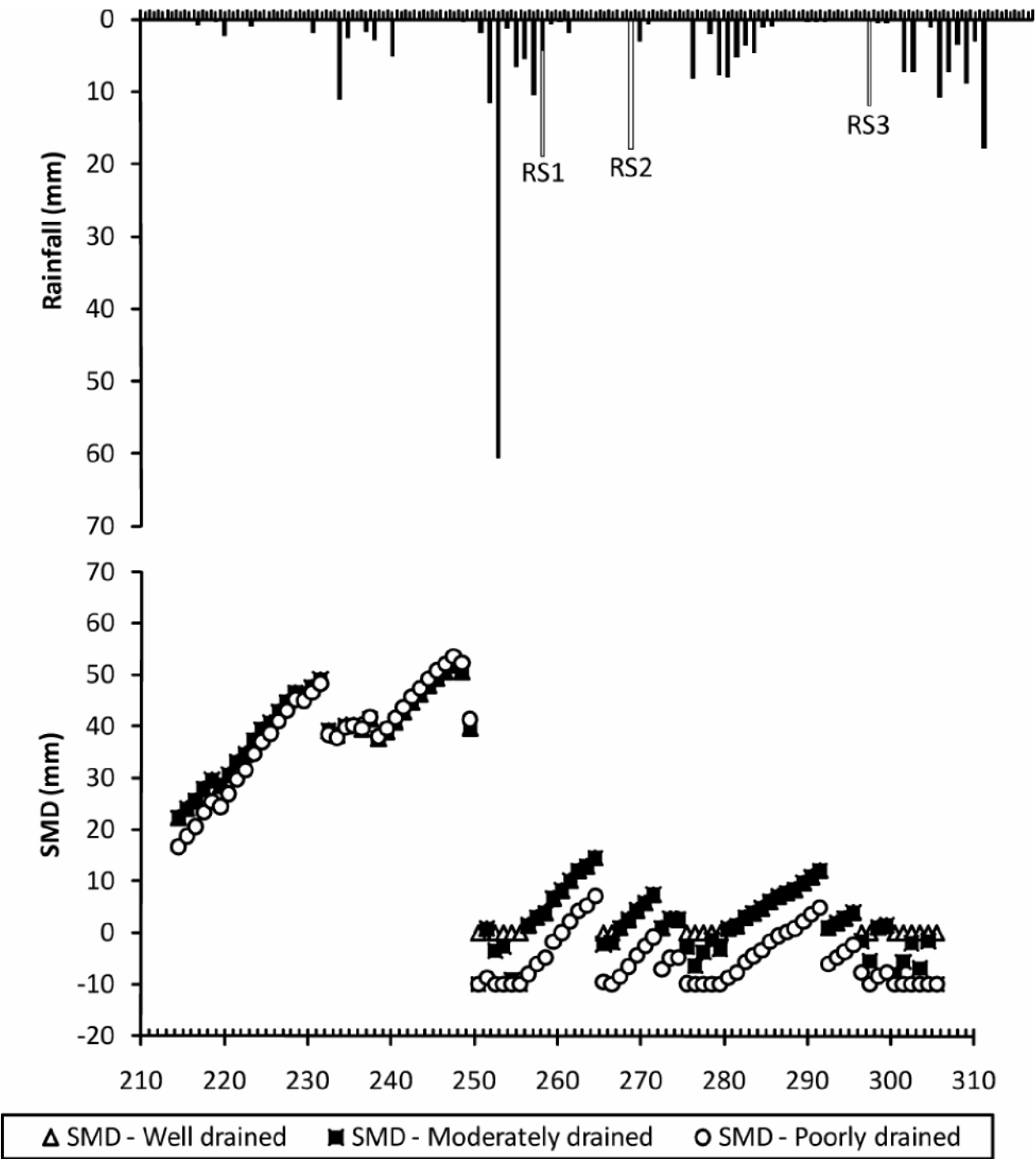
749 Fig 1 Map of study site showing ground elevation, topography, slope, soil conductivity,
750 groundwater flow direction, location of subplots, piezometers and diagram of runoff collection
751 channel and plot isolation.



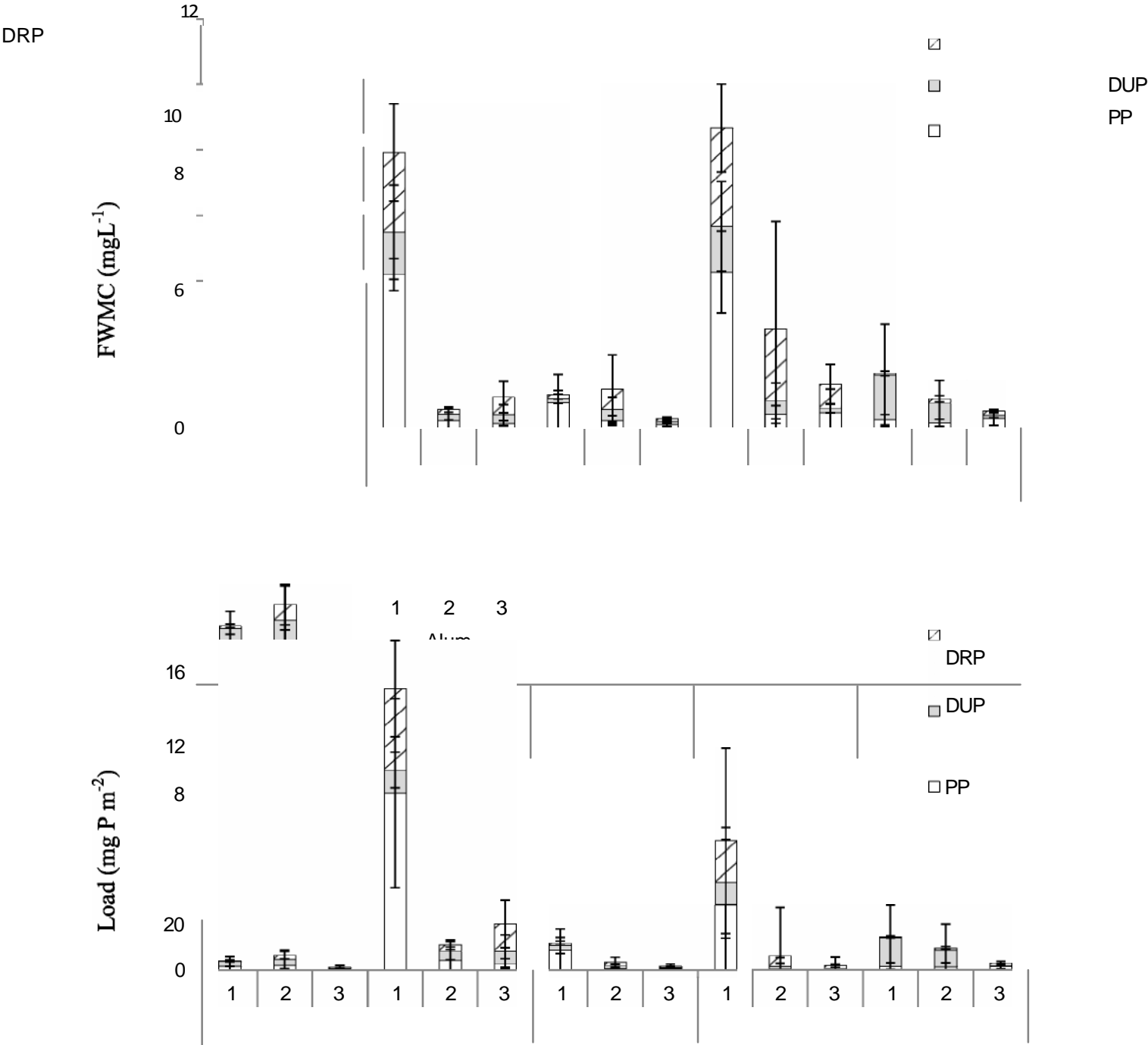
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754 Fig 2 Measured daily rainfall (mm) and simulated soil moisture deficit (SMD) for well, moderate
755 and poorly drained soils. Rainfall applied to plots during RS1-3 is added to measured daily
756 rainfall and used for simulated SMD calculation. X axis is in Julian Days.



759 Fig 3 Flow-weighted mean concentration and total loads of particulate phosphorus (PP),
 760 dissolved un-reactive phosphorus (DUP) and dissolved reactive phosphorus (DRP).



Soil

Control

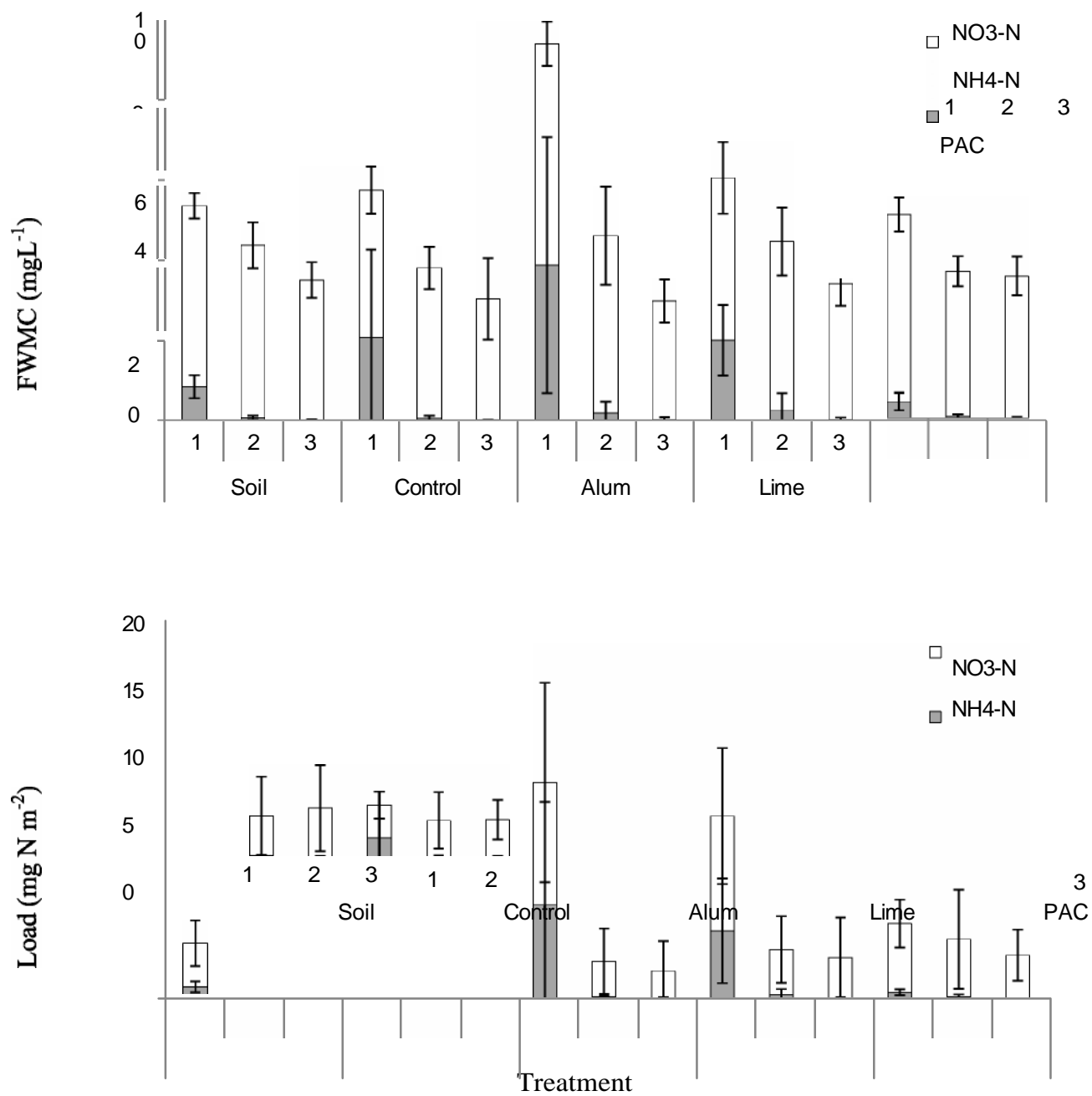
Alum

Lime

PAC

Treatment

762 Fig 4 Flow-weighted mean concentration and total loads of nitrate ($\text{NO}_3\text{-N}$) and ammonium
 763 ($\text{NH}_4\text{-N}$).



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765

766 Fig 5 Average rainfall intensity, runoff volume, time to runoff and soil volumetric water content
 767 for the first (RS1), second (RS2) and third (RS3) rainfall events.

768

769 Mean (average value for all plots)

